

Steady-state crystallization of Rydberg excitations in an optically driven lattice gas

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We study resonant optical excitations of atoms in a one-dimensional lattice to the Rydberg states interacting via the van der Waals potential which suppresses simultaneous excitation of neighboring atoms. Considering two- and three-level excitation schemes, we analyze the dynamics and stationary state of the continuously-driven, dissipative many-body system employing time-dependent density-matrix renormalization group (t-DMRG) simulations. We show that two-level atoms can exhibit only nearest neighbor correlations, while three-level atoms under dark-state resonant driving can develop finite-range crystalline order of Rydberg excitations. We present an approximate rate equation model whose analytic solution yields qualitative understanding of the numerical results.

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Strong, long-range interaction between Rydberg atoms [1] have positioned them as promising systems for quantum information processing [2–4], which motivated considerable experimental progress in preparing and studying such systems [5–12]. Rydberg atoms are also interesting in the context of many-body physics: It was predicted that the long-range interaction leads to spontaneous symmetry breaking and crystalline order in a continuous gas [13–15], formation of three-dimensional super-solids [16], as well as to vortex lattices [17] or fractional quantum Hall states [18] in two-dimensional (2D) systems with artificial magnetic fields. Coupling Rydberg atoms to light can give rise to novel photonic states with highly non-classical correlations [19, 20] and unusual nonlinear spectroscopic features [21, 22].

For regular arrays of coherently driven atoms, depending on the strength and detuning of driving lasers, different ground-state phases with crystalline order emerge [23–25]. Rydberg-dressed atoms [16, 26–28], i.e. ground-state atoms with a small admixture of Rydberg states, can also form crystalline structures with fractional fillings [29]. In the optically driven lattice gas [30], where the number of Rydberg excitations is not conserved, the preparation of the ground state of the system requires careful consideration and an adiabatic preparation scheme has been proposed and analyzed in [31]. A more natural approach to the formation of crystalline order of Rydberg excitations is to utilize the *stationary state* of a dissipative many-body system, which results from the interplay between continuous optical driving and spontaneous decay. Moreover, the steady state is an attractor of the system's dynamics and is therefore stable against small perturbations.

Here we study resonant optical excitations of Rydberg states of atoms using two- and three-level driving schemes. The van der Waals (vdW) interaction between the atoms leads to a Rydberg level shift and thereby blocks the excitation of an atom which is sufficiently close

to an already excited Rydberg atom. Specifically, we study a one-dimensional (1D) lattice with strong nearest neighbor interaction between the atoms, employing numerically exact t-DMRG simulations. We show that two-level driving leads to at most short-range correlations of Rydberg excitation probabilities of atoms at neighboring lattice sites. In contrast, for three-level atoms under the dark-state resonant driving, we find longer-range correlations and quasi-crystallization extending over several lattice periods. We derive an effective rate equations model, which can be solved exactly for the steady state yielding an expression for the correlation length. Finally, we estimate the influence of the tails of the vdW potential inducing small level shifts experienced by the next to nearest neighbor atoms, which give rise to an upper limit of the correlation length.

We thus consider a chain of N atoms trapped in a 1D lattice potential of period a [Fig. 1(a)] and examine one- and two-photon resonant excitations of atoms from the ground state $|g\rangle$ to the Rydberg state $|r\rangle$ [Fig. 1(b)]. In

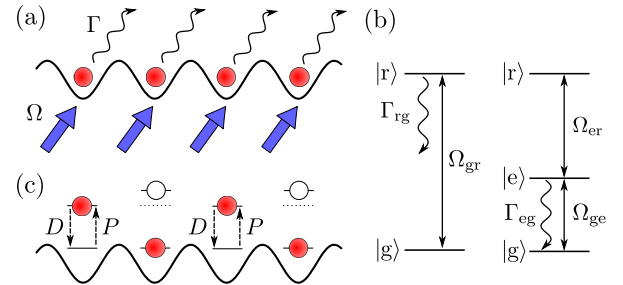


FIG. 1. (Color online). (a) Schematics of optically driven atoms in a lattice. (b) Starting from the ground state $|g\rangle$, the atoms are resonantly excited to the Rydberg state $|r\rangle$ either directly (left) or via resonant intermediate state $|e\rangle$ (right). (c) Illustration of the rate equations model with Rydberg blockade of neighboring atoms.

the two-level scheme, the transition $|g\rangle \rightarrow |r\rangle$ is driven by a laser field of (effective) Rabi frequency Ω_{gr} . In the three-level scheme, the Rydberg state $|r\rangle$ is populated from the ground state $|g\rangle$ via resonant intermediate state $|e\rangle$ in the coherent population trapping (CPT) or dark-state resonance configuration [32] with Rabi frequencies $\Omega_{ge} \gtrsim \Omega_{er}$. The corresponding atom-field interaction Hamiltonians are given, respectively, by $\mathcal{V}_2^j = -\hbar(\Omega_{gr}\hat{\sigma}_{rg}^j + \text{H.c.})$ and $\mathcal{V}_3^j = -\hbar(\Omega_{ge}\hat{\sigma}_{eg}^j + \Omega_{er}\hat{\sigma}_{re}^j + \text{H.c.})$, where $\hat{\sigma}_{\mu\nu}^j \equiv |\mu\rangle_{jj}\langle\nu|$ are the transition operators for atom j .

Spontaneous decay is described by Liouvillian terms in the equation of motion for the density operator ρ : $\mathcal{L}^j\rho = \frac{1}{2}(2\hat{L}^j\rho\hat{L}^{j\dagger} - \{\hat{L}^{j\dagger}\hat{L}^j, \rho\})$, where \hat{L}^j are the Lindblad generators. For the two-level scheme, $\hat{L}_2^j = \sqrt{\Gamma_{rg}}\hat{\sigma}_{gr}^j$ with Γ_{rg} being the (population) decay rate of Rydberg state $|r\rangle$. Although Γ_{rg} is typically small ($\sim 10^4$ Hz), it may be comparable to the Rabi frequency Ω_{gr} associated with either direct one-photon (UV) transition $|g\rangle \rightarrow |r\rangle$ with small dipole matrix element, or two-photon transition via far off-resonant intermediate states. For the three-level CPT scheme, we take into account only the large spontaneous decay rate Γ_{eg} ($\sim 10^7$ Hz) of the intermediate excited state $|e\rangle$ via $\hat{L}_3^j = \sqrt{\Gamma_{eg}}\hat{\sigma}_{ge}^j$; the decay rate Γ_{re} of Rydberg state can safely be neglected in comparison with Ω_{er} .

For a single (isolated) two- or three-level atom under continuous driving, the steady-state population of the Rydberg state is given, respectively, by

$$\langle\hat{\sigma}_{rr}\rangle \approx \frac{|\Omega_{gr}|^2}{2|\Omega_{gr}|^2 + \gamma_{rg}^2 + \Delta^2}, \quad (1a)$$

$$\langle\hat{\sigma}_{rr}\rangle \approx \frac{|\Omega_{ge}|^2(|\Omega_{ge}|^2 + |\Omega_{er}|^2)}{(|\Omega_{ge}|^2 + |\Omega_{er}|^2)^2 + (\gamma_{eg}^2 + 2|\Omega_{ge}|^2)\Delta^2}, \quad (1b)$$

where $\gamma_{\mu\nu} = \frac{1}{2}\Gamma_{\mu\nu}$ and Δ is the one- or two-photon detuning of the laser frequency from the $|g\rangle \rightarrow |r\rangle$ transition resonance. It follows from Eqs. (1) that the Rydberg state population $\langle\hat{\sigma}_{rr}\rangle$ of an atom is a Lorentzian function of Δ , with the width $w = \sqrt{2|\Omega_{gr}|^2 + \gamma_{rg}^2}$ for the direct excitation and $w = (|\Omega_{ge}|^2 + |\Omega_{er}|^2)/\sqrt{\gamma_{eg}^2 + 2|\Omega_{ge}|^2}$ for the CPT excitation schemes.

Finally, pairs of atoms i and j interact with each other via the vdW potential [33] $\mathcal{V}_{\text{vdW}}^{ij} = \hbar\hat{\sigma}_{rr}^i \frac{C_6}{d_{ij}^6} \hat{\sigma}_{rr}^j$, where $d_{ij} = a|i-j|$ is the interatomic distance. Once an atom i is excited to the Rydberg state $|r\rangle$, it shifts the atom j out of the resonance by $\Delta = U/|i-j|^6$, where $U \equiv C_6/a^6$. The excitation of atom j to state $|r\rangle$ is then blocked if $\Delta \gtrsim w$, which determines the blockade distance $d_b \simeq \sqrt[6]{C_6/w}$. Throughout this paper, we assume that $a < d_b < 2a$, so that there is a Rydberg blockade only between the neighboring atoms.

The density matrix ρ of the system of N atoms obeys

the master equation

$$\dot{\rho} = -\frac{i}{\hbar}[\mathcal{H}_l, \rho] + \mathcal{L}_l\rho, \quad (2)$$

with the Hamiltonian $\mathcal{H}_l = \sum_j \mathcal{V}_l^j + \sum_{i<j} \mathcal{V}_{\text{vdW}}^{ij}$ and the Liouvillian $\mathcal{L}_l\rho = \sum_j \mathcal{L}_l^j\rho$ for $l = 2$ or 3 level atoms.

Since the interaction of an atom with its immediate neighbor is much stronger than that with the atoms further apart, we truncate the vdW potential $\mathcal{V}_{\text{vdW}}^{ij}$ to nearest neighbor (NN), $\mathcal{V}_{\text{NN}}^{ij} = \hbar\hat{\sigma}_{rr}^i U \hat{\sigma}_{rr}^j$ for $i = j-1$ and $\mathcal{V}_{\text{NN}}^{ij} = 0$ otherwise. We have performed exact numerical integrations of Eq. (2) for small systems of several ($N \leq 7$) atoms interacting via the \mathcal{V}_{vdW} and \mathcal{V}_{NN} potentials, and verified that they yield similar results for both two- and three-level excitation schemes. We will therefore employ from now on the \mathcal{V}_{NN} potential, returning to the corrections due to longer-range interactions of the \mathcal{V}_{vdW} potential later on.

We obtain the time evolution and the steady state of the full many-body density matrix Eq. (2) employing the t-DMRG method [34, 35]. Our implementation follows the original proposal by Vidal [36], generalized to open quantum systems [37, 38]. Simulations for up to $N \sim 10^2$ atoms are possible as the relaxation keeps the entanglement inside the system small. As a consequence, the usual limitations concerning the propagation time do not apply and the t-DMRG integration can be performed for, in principle, arbitrarily long times. Furthermore, since the stationary state is an attractor of the dynamics of the system, accumulated errors self-correct. We verified that for small systems the exact and t-DMRG solutions with the \mathcal{V}_{NN} potential are indistinguishable.

Results of the t-DMRG simulations for two- and three-level atoms in a realistically large lattice of length $30a$ with open boundary conditions are shown in Fig. 2. Since interactions between the atoms suppress the Rydberg excitations, the atoms at the boundaries $j = 0$ and $j = N-1$ having only one neighbor acquire the largest population of the Rydberg state $|r\rangle$. We also observe period-2 spatial oscillations of excitation probabilities $\langle\hat{\sigma}_{rr}^j\rangle$. For two-level atoms, the amplitude of these oscillations, as well as the average Rydberg state population is low, both for weak $\Omega_{gr} < \Gamma_{rg}$ and moderately strong $\Omega_{gr} \simeq \Gamma_{rg}$ driving, Fig. 2(a) and (b). This is due to the fact that even for a noninteracting two-level atom under strong driving $\Omega_{gr} \gg \Gamma_{rg}$ the excited state population saturates to $\langle\hat{\sigma}_{rr}\rangle \rightarrow \frac{1}{2}$ [cf. Eq. (1a)]. In contrast, for a (noninteracting) three-level atom under the CPT excitation scheme, the population of Rydberg state can be very large, $\langle\hat{\sigma}_{rr}\rangle \rightarrow 1$, when $\Omega_{ge} \gg \Omega_{er}$ [cf. Eq. (1b)]. For a chain of strongly interacting atoms, we then observe a large amplitude of spatial oscillations of the Rydberg excitation probabilities $\langle\hat{\sigma}_{rr}^j\rangle$ extending over many lattice sites, Fig. 2(c).

Atoms excited to Rydberg states are typically no longer trapped in an optical lattice, although there has

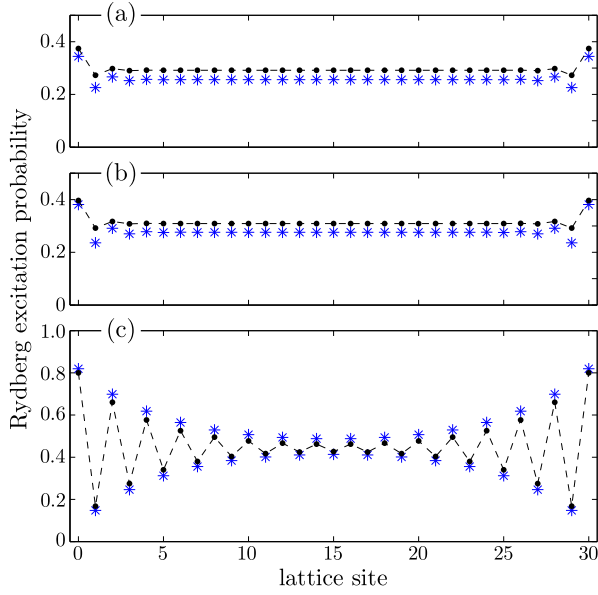


FIG. 2. (Color online). Steady-state Rydberg excitation probabilities $\langle \hat{\sigma}_{rr}^j \rangle$ of two- and three-level atoms in a lattice of length $30a$ ($N = 31$ atoms). Black dots (connected by dashed lines) show the t-DMRG solutions of Eq. (2) with the truncated potential \mathcal{V}_{NN}^{ij} , and blue stars are the REs solutions for the hard-core (infinite NN) potential. For the two-level atoms, the parameters are $U = 2\Omega_{gr}$, $\Gamma_{rg} = \Omega_{gr}$ (a) and $\Gamma_{rg} = \frac{1}{4}\Omega_{gr}$ (b). For the three-level atoms, $U = 2\Omega_{ge}$, $\Gamma_{eg} = 4\Omega_{ge}$, and $\Omega_{er} = \frac{1}{5}\Omega_{ge}$ (c).

been some progress in trapping Rydberg atoms using ponderomotive potentials [39]. An important question is therefore, whether the steady-state crystalline order of Rydberg excitations can be reached during times short enough for the center-of-mass motion of the excited atoms to be negligible. In Fig. 3 we show the time evolution of Rydberg state populations of atoms initially in the ground state $|g\rangle$. Note that applying the driving lasers to all the atoms at once yields the global stationary state of the system after times two to three orders of magnitude longer than the steady-state equilibration time of an isolated atom. The reason for this is the initial formation of finite-size anti-ferromagnetic domains with dislocation defects in between, which require excessive healing times. But the steady-state is independent of the initial conditions and details of preparation, therefore faster schemes for preparing the global stationary state of the system may exist. Similar to classical crystals, it is indeed much faster to “grow” the Rydberg quasi-crystal by first applying the lasers to the atoms at the lattice boundary and then gradually extending the irradiated region until the full Hamiltonian \mathcal{H}_3 is realized. In Fig. 3 we verify this intuitive approach. Since the crystalline order is rooted in the Rydberg blockade of the NN sites, a “sweep” velocity as fast as one lattice period a per single-atom equilibration time $\Gamma_{eg}/(\Omega_{ge}\Omega_{er})$ can be applied to reach the

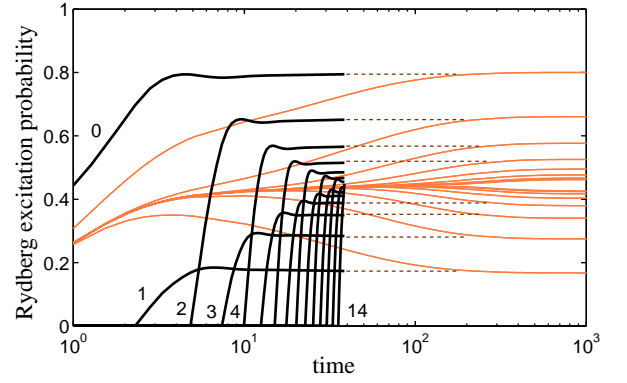


FIG. 3. (Color online). Dynamics of Rydberg excitation probabilities for simultaneous application of driving lasers at time $t = 0$ to all the atoms (thin red lines), and for sequential application—“sweep”—of the lasers to atoms $j = 0, 1, 2, \dots$ (thick black lines). All parameters are as in Fig. 2(c) and time is in units of $\frac{\Gamma_{eg}}{\Omega_{ge}\Omega_{er}}$.

steady-state configuration.

We now describe an effective rate equations (REs) model, which yields an analytic solution for the stationary state of the system. We are interested in the Rydberg excitation probabilities $\langle \hat{\sigma}_{rr}^j \rangle$ of atoms and their steady-state correlations $\langle \hat{\sigma}_{rr}^j \hat{\sigma}_{rr}^k \rangle$, which allows us to disregard the coherences between the atoms. We restrict the interatomic interactions to the complete blockade of Rydberg excitations of NNs, assuming the potential \mathcal{V}_{NN}^{ij} with $U \rightarrow \infty$. At each lattice site, we then have two incoherent processes: a state dependent pump with rate P and a deexcitation with rate D ,

$$\hat{L}_p^j = \sqrt{P} (\hat{\sigma}_{rr}^{j-1} - \mathbb{1}) \hat{\sigma}_{rg}^j (\hat{\sigma}_{rr}^{j+1} - \mathbb{1}), \quad (3a)$$

$$\hat{L}_d^j = \sqrt{D} \hat{\sigma}_{gr}^j. \quad (3b)$$

The ratios of the two rates $\kappa_l \equiv \frac{P}{D} = \frac{1 - \langle \hat{\sigma}_{rr} \rangle}{\langle \hat{\sigma}_{rr} \rangle}$ for the $l = 2$ and 3 level atoms are obtained from Eqs. (1) as

$$\kappa_2 = \frac{|\Omega_{gr}|^2}{|\Omega_{gr}|^2 + \gamma_{rg}^2}, \quad \kappa_3 = \frac{|\Omega_{ge}|^2}{|\Omega_{er}|^2}.$$

In this model, the density matrix $\rho(t)$ obeys the equation of motion

$$\dot{\rho} = \sum_j (2\hat{L}_p^j \rho \hat{L}_p^{j\dagger} - \{\hat{L}_p^{j\dagger} \hat{L}_p^j, \rho\} + 2\hat{L}_d^j \rho \hat{L}_d^{j\dagger} - \{\hat{L}_d^{j\dagger} \hat{L}_d^j, \rho\}). \quad (4)$$

After sufficient relaxation time, the density matrix attains an essentially classical form $\rho(t) = \sum_{\{n_j\}} p(\{n_j\})(t) |\{n_j\}\rangle \langle \{n_j\}|$, where $p(\{n_j\})$ is the probability of configuration $\{n_j\} \in (0, 1)^N$. Classical state-space dimension grows exponentially with N , precluding numeric integration for large systems. However,

the steady state of REs fulfills the detailed balance relation

$$\frac{p(\{m_j\})}{p(\{n_j\})} = \kappa_l^{\sum_j (m_j - n_j)}. \quad (5)$$

States with the same number of excitations have equal weight and the partition function is given by $Z_N = \sum_{M=0}^N \Omega(M, N) \kappa_l^M$, where $\Omega(M, N)$ is the number of possible arrangements of M excitations on a lattice of N sites. In 1D, we have the analytic expression $\Omega(M, N) = \binom{N-M+1}{N}$, which enables efficient calculation of all the steady state probabilities $p(\{n_j\})$.

In Fig. 2 we compare the solutions of the REs model with the results of t-DMRG simulations, observing reasonable agreement, especially for the three-level excitation scheme of the atoms. In all cases, however, the decay of correlations is correctly captured by the REs model.

For a large lattice $N \gg 1$, the partition function converges to $Z_N = \cosh\left((2+N)\frac{\sqrt{\kappa_l}}{2}\right)$, with which the Rydberg excitation probabilities at odd sites j (the boundary being at $j=0$) are given by $\langle \hat{\sigma}_{rr}^j \rangle = e^{-1/(2\sqrt{\kappa_l})}(1 + e^{-j/\sqrt{\kappa_l}})/2$. The decay of spacial oscillations of probabilities $\langle \hat{\sigma}_{rr}^j \rangle$ is characterized by the correlation length

$$\xi(\kappa_l) = \sqrt{\kappa_l} a. \quad (6)$$

Note that in a translationally invariant system (i.e. in the bulk), the excitation probability is uniform in space, but the onset of crystallization is revealed by the density-density correlations, which can also be calculated in the REs model. We find that $\xi(\kappa)$ is indeed the corresponding correlation length.

For the two-level excitation scheme of atoms, $\kappa_2 < 1$, we then obtain the correlation length $\xi < a$, i.e. shorter than the lattice spacing. Using a mean-field approximation for the same system in the stationary state, Lee *et al.* [25] predicted a phase transition to anti-ferromagnetic order (period-2 density wave). The reason for this discrepancy is that the mean field approximation is inadequate in 1D. We note that even in a 2D square lattice the phase transition to the antiferromagnetic state cannot be reached with the two-level atoms; the 2D extension of the REs approach leads to the classical hard-square model, for which a phase transition to the Néel ordered (checkerboard) phase occurs at the critical value of $\kappa_{\text{crit}} \approx 3.7962$ [40].

For the CPT excitation of three-level atoms, one can tune κ_3 by the Rabi frequencies Ω_{ge} and Ω_{er} , attaining large but finite correlation lengths $\xi > a$. Further increase of ξ is constrained by the validity of the NN interaction approximation. We therefore consider the corrections originating from the hitherto neglected interactions of the atom with the next nearest neighbors (NNNs). To estimate the maximal effect of the NNN interaction, let us assume a strong driving, resulting in Rydberg excitations at every other lattice site (half filling). A non-blocked atom is then likely to be detuned by $\Delta' = \Delta/2^6$

due to the interaction with the NNN Rydberg atoms. Expressing the NN detuning $\Delta = \beta w$ in terms of the excitation linewidth w and using Eqs. (1b), we obtain a modified

$$\kappa'_3 = \frac{|\Omega_{ge}|^2}{|\Omega_{er}|^2 + (|\Omega_{ge}|^2 + |\Omega_{er}|^2) \left(\frac{\beta}{64}\right)^2} \leq \left(\frac{64}{\beta}\right)^2. \quad (7)$$

To ensure almost perfect NN blockade, it is reasonable to take $\beta \approx 10$ and $\sim 1\%$ excitation probability for the blocked atom. In 1D, the correlation length is therefore limited to values of $\xi \lesssim 7a$, due to the softness of the vdW interaction potential and the long wings of the Lorentzian excitation profile. In 2D, the phase transition to Néel order may occur for three-level atoms, but since the effects of the NNN interactions will be more pronounced, the existence of a phase transition in 2D remains an open question. We finally note that for the dipole-dipole interaction $\mathcal{V}_{\text{DD}}^{ij} \propto \frac{C_3}{d_{ij}^3}$, Eq. (7) reduces to $\kappa'_3 \leq (8/\beta)^2$ leading to a short correlation length $\xi < a$.

To conclude, we have found that the collective steady-state of an ensemble of three-level atoms in a 1D lattice can exhibit quasi-crystallization of Rydberg excitations with a correlation length extending over many lattice periods. In contrast, for two-level atoms even under strong driving, the correlations are only between the neighboring atoms and the phase transition in 2D to Néel order cannot occur. Using the time-dependent DMRG simulations for several tens of atoms, we have shown that for uniform optical driving of all the atoms, long-range crystalline order of Rydberg excitations is attained only after very long times. A sequential excitation of neighboring atoms by dynamically “sweeping” the lattice with the driving laser, can result in the same steady-state of the system in a much shorter time given by the single-atom equilibration time multiplied by the number of sites. We derived an effective rate equations model whose exact steady-state solution, being in good agreement with the numerical calculations, yielded explicit analytic expressions for the Rydberg excitation probabilities and correlation length.

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